

LIQUID MIXING ON DISTILLATION PLATES

by

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INTRODUCTION

The design of distillation columns, based on the concept of the "theoretical plate" is a relatively well-developed art. A "theoretical plate" is a plate on which the contact between vapor and liquid is sufficiently good so that the vapor leaving the plate has the same composition as the vapor in equilibrium with the overflow from the plate (1).

Unfortunately actual plates do not perform as do theoretical plates, and it is desirable that expressions be available to represent the deviation from ideality. The relationship generally used is expressed as plate efficiency. Many different forms of the plate efficiency have been proposed, but the most commonly used is the "point" efficiency suggested by Murphree (2).

As the name implies, the "point" efficiency can be computed only for a region of the plate on which the liquid and the vapor phases are both of uniform composition, and then only when sufficiently reliable methods are available for predicting mass transfer rates. To date, the only cases in which plate (or overall) efficiency can be precisely related to point efficiency is when the liquid on the plate is completely mixed or when the liquid flows across the plate in plug flow. It has been shown by various investigators (3), (4), (5) that neither of these extreme flow regimes actually exist on distillation plates; the actual case lies somewhere between.

The magnitude of the plate efficiency, under conditions when entrainment is negligible, is therefore, affected not only by the

rate of mass transfer in the liquid and gas phases, but also by the degree of fluid mixing on the plate. As will be pointed out, the analysis of fluid interaction on a tray is well beyond exact mathematical and fluid-dynamical treatment. For this reason experimental measurement and modeling techniques have been exclusively employed to describe mixing on distillation plates and its effect on plate efficiency.

Various models, physical as well as mathematical, have been proposed to describe the liquid mixing on distillation plates. The pool model, which was originally proposed by Kirchbaum (6) and later revived by Nord (7) and Gatreaux and O'Connell (8), postulates that the tray may be considered as a series of completely mixed pools along the length of the tray. While it is assumed that there is complete mixing within each pool, there is no mixing between pools. A plate with a single pool corresponds to a perfectly mixed plate and one with an infinite number of pools to an unmixed or plug flow plate.

The recycle models of Oliver and Watson (9) and Warzel (10) assume that liquid mixing is effected by a recycle of liquid which moves from the tray exit to the tray inlet. The parameter which characterizes mixing is the concentration jump at the inlet weir.

Splashing models have been proposed by Johnson and Marangozis (11) and Crozier (12). In these models, splashing of the liquid is considered to be the major mixing process.

The axial dispersed plug flow model has been applied to distillation trays or other flow systems by Anderson (13), Wehner

and Wilhelm (14), Gerster et. al. (15) and others (16), (17), (5). This model assumes that the rate of mixing or dispersion of a component is proportional to the concentration gradient of that component.

Foss (18) has used the residence-time concept to characterize the degree of liquid mixing on distillation trays. He has shown how the residence time distribution function may be used to include liquid mixing effects in the calculation of plate efficiencies.

Strand (19) has proposed a model which considers the distillation plate to be an axial dispersed plug flow vessel around which a fraction of the entering liquid stream is effectively bypassed. He has shown that the bubble tray design method proposed by the American Institute of Chemical Engineers (20) could be substantially improved by including a liquid bypassing effect. Strand also suggests other possible changes in the A.I.Ch.E. method. One possible change would be to consider the tray to be non-uniform, both in the contacting achieved on various parts of the tray and in the degree of liquid mixing. He points out that the dispersion coefficient data used in the development of the A.I.Ch.E. method were obtained in a long narrow tray section so that variations transverse to the liquid path were negligible.

This report is not concerned with plate efficiencies as such. It is concerned with the fluid flow behavior in general and the liquid mixing and dispersion in particular, which takes place on a distillation plate. It would be expected that the flow behavior would be affected by such factors as liquid and

vapor flow rates, tray design, and the physical properties of the fluids.

Various models which may be used to describe flow behavior are discussed, among them, the dispersion model, the perfectly mixed tanks in series model, mixed models, and the Γ -distribution model. Experimental methods for determining the parameters of these models are given. One such method is the pulse testing technique which has been discussed by several authors (21), (22), but never actually used for studies of distillation trays.

A proposed investigation of liquid mixing and dispersion on distillation plates is outlined. It is suggested that point age distributions as well as residence time distributions be experimentally determined, the latter by the use of pulse testing techniques.

Many authors have pointed out the need for an investigation of the type proposed in this report (3), (5), (19), (23), (4). In the recent report of the Workshop on Automatic Control Research to the Control Advisory Committee of the American Automatic Control Council and the National Science Foundation (24) the following statements were made concerning the formulation and use of theoretical mathematical models of chemical processes.

Automatic data reduction techniques with computers have made it relatively easy to develop overall transfer function or performance function models relating a particular process output variable to a particular input variable change. This is especially true for pulse type inputs. Because of the special data reduction problems involved, higher-order effects may be lost or at best very difficult to obtain. The following questions regarding these studies remain relatively unanswered as of this time:

1. What are the indispensable dependent variables of any particular process? That is, just how many separate transfer functions are necessary to describe the system adequately? What are the best data presentation methods for expressing this data?
2. Just as in the case of linearized small-perturbation type simulations what are the limits of input variable manipulation for which the linear transfer function representation is adequate?
3. What are the criteria to be followed in deciding whether higher-order effects may or may not be important in any specific case?

Concerning the area of process dynamics, the report continues:

Some general observations regarding the problem areas in process dynamics can also be made at this time. These are:

1. Most difficulties in mathematical model making resolve themselves directly to the characterization of fluid flow phenomena occurring in the process such as turbulence, liquid mixing, etc.
2. Control in chemical processing almost always involves the manipulation of the flow of a fluid stream (here fluid may mean gas, liquid, or fluidized solid.)

Hence fluid flow problems are at the heart of all our control investigations, while at the same time being the process dynamic phenomena which we least understand.

BASIC NOTIONS OF FLUID DISPERSION

The degree to which the aerated liquid mass flowing across a distillation plate is mixed or dispersed may be characterized by either the distribution of residence times of the liquid elements flowing across the plate or the concentration profile of the liquid on the plate (15). From a theoretical viewpoint, both of these relationships may be obtained analytically.

Consider a single phase, multicomponent fluid system. A full description of this system is given by the following relationships:

$$\frac{\partial C_1}{\partial t} = - (\nabla \cdot \{C_1 \bar{u} + \bar{J}_1\}) + R_1 \quad i = 1, 2, \dots, n \quad (1)$$

$$\frac{\partial \rho \bar{u}}{\partial t} = - [\nabla \cdot \{\rho \bar{u} \bar{u} + \bar{\pi}\}] + \sum_{i=1}^n \rho_i \bar{g}_i \quad (2)$$

$$\frac{\partial}{\partial t} \left\{ \hat{U} + \frac{1}{2} u^2 \right\} = - (\nabla \cdot \{ \rho (\hat{U} + \frac{1}{2} u^2) \bar{u} + \bar{q} + [\bar{\pi} \cdot \bar{u}] \}) \quad (3)$$

Equation (1) is the equation of continuity, a mass balance.

Equation (2) is a momentum balance, variously called the equation of motion or the Navier-Stokes equation.

Equation (3) is an energy balance. These Equations describe the behavior of a fluid system in general within the framework of mechanics of continuous media (25). They are seldom used in the complete form given here since the exact solution of these coupled equations is beyond the scope of present day mathematics. The normal procedure is to disregard terms that are physically

negligible or identically zero, thereby obtaining simpler equations for a given situation. For example, consider an isothermal, Newtonian, incompressible fluid of constant mass density and viscosity. Equation (3) is no longer needed and equations (1) and (2) become

$$\frac{\partial C_i}{\partial t} = -\bar{u} \nabla \cdot C_i - \nabla \cdot \bar{J}_i + R_i \quad (4)$$

$$\rho \frac{D\bar{u}}{Dt} = -\nabla p - [\nabla \cdot \bar{\tau}] + \rho \bar{g} \quad (5)$$

For a given physical situation of simple geometry (e.g. flow in a circular tube) equations (4) and (5) are more readily solved for laminar flow. This is because in laminar flow the molar flux \bar{J}_i and the stress tensor $\bar{\tau}$ are expressed in terms of Fick's law of diffusion and Newton's law of viscosity. When considering turbulent flow \bar{J}_i and $\bar{\tau}$ are given by (26)

$$\bar{J}_i = \bar{J}_i^{(1)} + J_i^{(t)} \quad (6)$$

$$\bar{\tau} = \bar{\tau}^{(1)} + \bar{\tau}^{(t)} \quad (7)$$

The superscripts (1) and (t) refer to laminar and turbulent contributions respectively. The $\bar{J}_i^{(1)}$ and $\bar{\tau}^{(1)}$ can be given by the same expressions as for purely laminar flow. The difficulty here is that only semiempirical expressions are available for $J_i^{(t)}$ and $\bar{\tau}^{(t)}$.

If the geometry of the physical system becomes complicated even the laminar flow situation is impossible, or at best, very difficult to solve.

It is therefore convenient to treat fluid mixing from semi-empirical and/or mathematical or physical modeling techniques. Furthermore, it is essential to confirm results from such a treatment experimentally.

Types of Flow

Two distinct types of mixing can be considered in continuous flow systems (27). One is macromixing, i.e., the primary mixing causing the non-uniform residence time distribution of elements of a fluid passing through the system. The other is micromixing, i.e., mixing on a molecular scale. This distinction becomes important when considering systems where non-linear rate processes are taking place. Since most of the mass transfer processes are of first order, or can be approximately considered to be first order, mixing on a distillation plate may be treated from either point of view.

Two ideal flow patterns which are often used to approximate real systems are plug flow and complete mixing (21). Patterns of flow other than plug or complete mixing flow may be called non-ideal flow patterns (21). Terms such as channelling, eddying, recycling, and flow with dead space or stagnant pockets are used to describe non-ideal flow. These descriptive terms are not completely mutually exclusive.

Most of the flow systems encountered are either single or two phase systems. According to Leonard (3), the two phase system may be viewed as a complication of the one phase system. The kinetic energy of the second phase offers an additional

source of mixing energy as well as an additional impediment to flow of the first phase. When describing the flow behavior of a two phase system in which there is a first order or linear rate process taking place, it is possible to treat each phase independently. If a non-linear rate process is taking place, independent treatment of the phases may no longer be possible (27).

Open and Closed Vessels

As discussed by Levenspiel (21) a closed vessel is defined as one for which fluid enters and leaves by bulk flow alone. Plug flow exists in the entrance and exit streams, i.e., diffusion and dispersion are absent at entrance and exit.

An open vessel is one where neither the entrance or exit streams satisfy the plug flow requirements of the closed vessel. When either the entrance or exit stream alone satisfies the closed vessel requirements, the vessel is said to be closed-open or open-closed.

Age Distributions

To be able to account exactly for non-ideal flow requires knowledge of the complete flow pattern of the fluid in the vessel. Due to the difficulties associated with obtaining and interpreting such information, an alternate approach is used which requires knowledge only of how long different elements of fluid reside in the vessel. This partial information is

relatively simple to obtain experimentally, can be easily interpreted, and either with or without flow models, yields information which in many cases allows a satisfactory accounting of the non-ideal flow.

The experimental technique used for finding this desired distribution of residence times of fluid elements in the vessel is a stimulus-response technique using tracer in the flowing fluid. The input signal is a tracer introduced in a known manner into the fluid stream entering the vessel. This input signal may be of any type. The response or output signal is then the recording of tracer concentration at a point within the vessel or in the outlet stream.

Before discussing age distributions, it is desirable to define the mean residence time and reduced time. The mean residence time, \bar{t} , of fluid in a vessel is defined as

$$\bar{t} = \frac{V}{\dot{V}} = \frac{\text{Volume of the vessel available for flow}}{\text{Volumetric flow rate of fluid through the vessel}} \quad (8)$$

Reduced time, θ , is defined as

$$\theta = \frac{t}{\bar{t}} \quad (9)$$

The discussion which follows is based in part on the work of Levenspiel (21) and De Maria and Longfield (28).

Internal age distribution. The vessel contains, in general, fluid elements of varying ages; ages being the times the fluid elements have spent in the vessel. Let $\underline{I}(\theta)$ be the internal age distribution function, which is defined so that $\underline{I}(\theta)d\theta$ is the fraction of fluid elements with ages between θ and $\theta + d\theta$.

A typical plot of $\underline{I}(\theta)$ versus θ is given in Fig. 1. It follows that the area under the curve is one.

$$\int_0^{\infty} \underline{I}(\theta) d\theta = 1 \quad (10)$$

The fraction the fluid with ages less than θ_1 is shown in Fig. 1 as the shaded area and is given by

$$\int_0^{\theta_1} \underline{I}(\theta) d\theta \quad (11)$$

the fraction of fluid with ages greater than θ_1 is

$$\int_{\theta_1}^{\infty} \underline{I}(\theta) d\theta = 1 - \int_0^{\theta_1} \underline{I}(\theta) d\theta \quad (12)$$

The internal age distribution $\underline{I}(t)$ based on time rather than reduced time is related to $\underline{I}(\theta)$ as

$$\underline{I}(\theta) = \bar{t} \underline{I}(t) \text{ with } \int_0^{\infty} \underline{I}(t) dt = 1 \quad (13)$$

Exit age distribution. In a manner similar to $\underline{I}(\theta)$, let $\underline{E}(\theta)$ denote the distribution of ages of all fluid elements leaving the vessel. $\underline{E}(\theta)$ is defined so that $\underline{E}(\theta)d\theta$ is the fraction of material in the exit stream between the ages of θ and $\theta + d\theta$.

It follows that

$$\int_0^{\infty} \underline{E}(\theta) d\theta = 1 \quad (14)$$

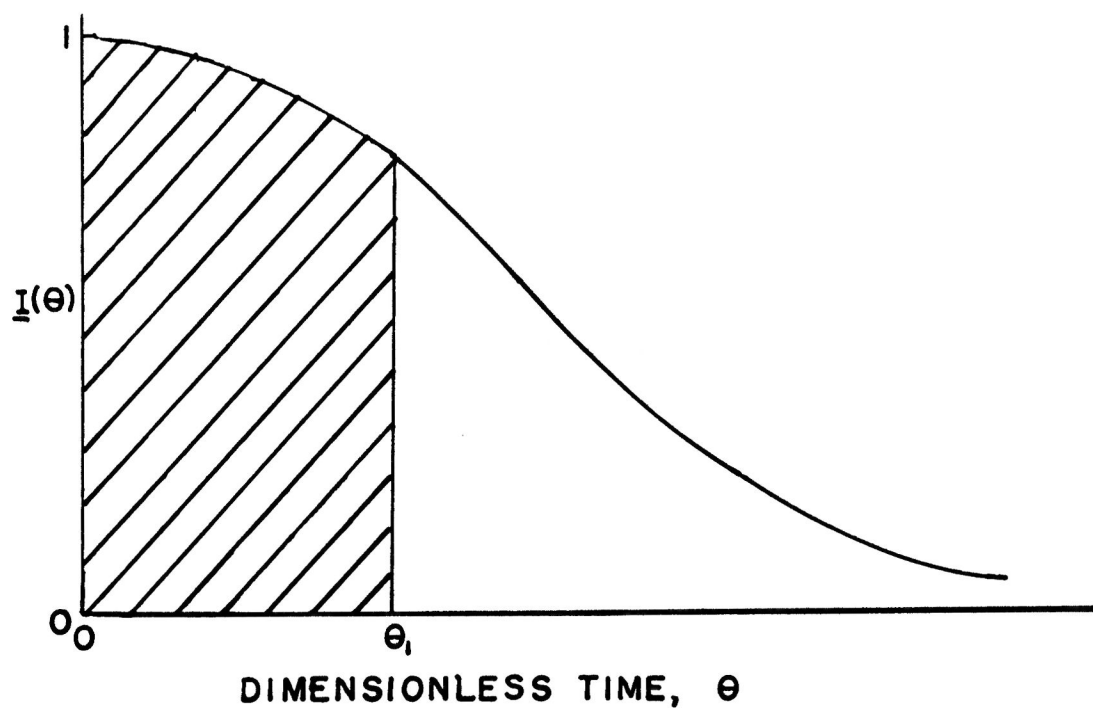


FIG. 1. TYPICAL INTERNAL AGE DISTRIBUTION CURVE

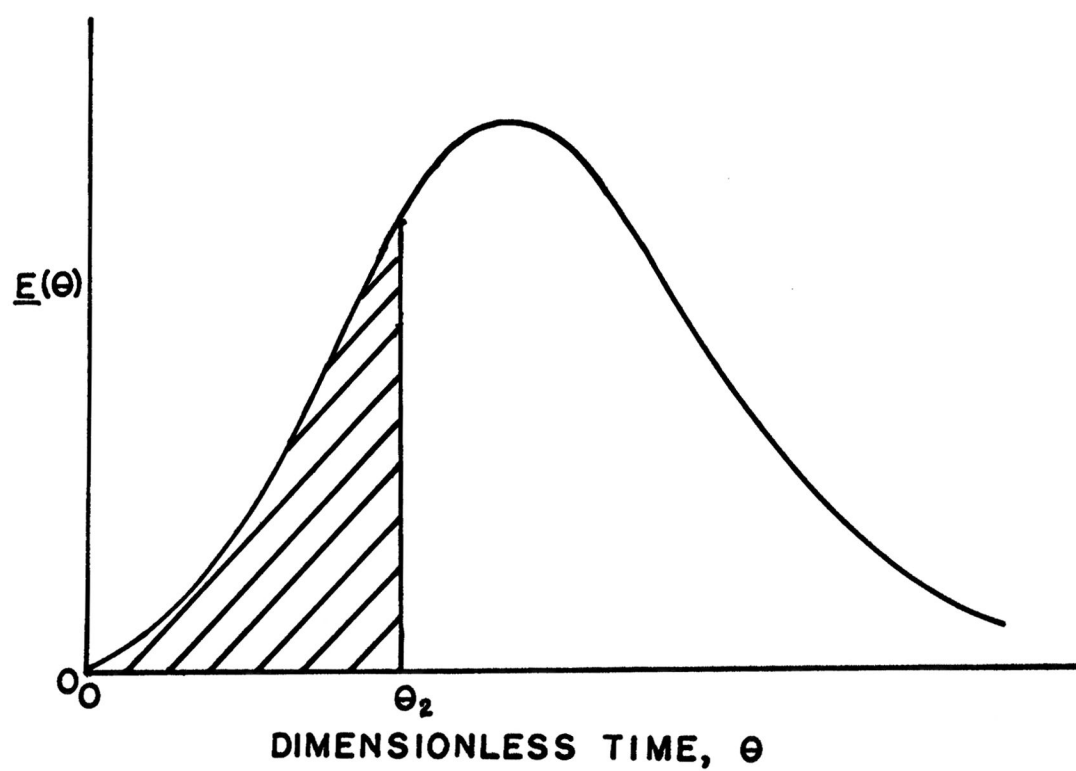


FIG. 2. TYPICAL EXIT AGE DISTRIBUTION CURVE

a typical $\underline{E}(\theta)$ curve is given in Fig. 2. The fraction of material in the exit stream younger than age θ_2 , the shaded area of Fig. 2 is

$$\int_0^{\theta_2} \underline{E}(\theta) d\theta \quad (15)$$

and the fraction older than θ_2 is

$$\int_{\theta_2}^{\infty} \underline{E}(\theta) d\theta = 1 - \int_0^{\theta_2} \underline{E}(\theta) d\theta \quad (16)$$

$\underline{E}(\theta)$ is variously referred to as the exit age distribution function, the exit residence time distribution or simply the residence time distribution function (r.t.d.f.). If time, t , is used instead of θ ,

$$\underline{E}(\theta) = \tau \underline{E}(t) \text{ with } \int_0^{\infty} \underline{E}(t) dt = 1 \quad (17)$$

The F-curve. With no tracer initially present, let a step function in time of tracer be introduced into the fluid entering the vessel in such a manner that the volumetric flow rate to the vessel remains constant. Then the concentration-time curve for tracer in the exit fluid stream, measured in terms of tracer concentration in the entering stream, C_0 , and reduced time θ is called the F-curve. As shown in Fig. 3, the range of F is $0 \leq F \leq 1$.

The C-curve. The curve which describes the concentration-time function of tracer in the exit stream of a vessel in

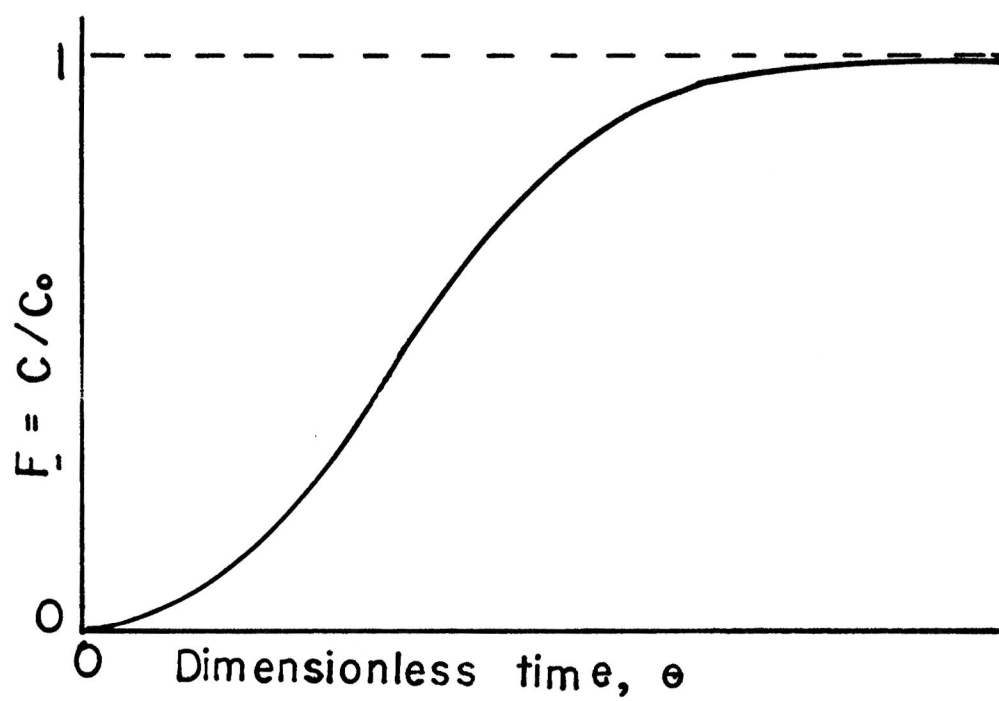


Fig.3. Typical \underline{F} -curve

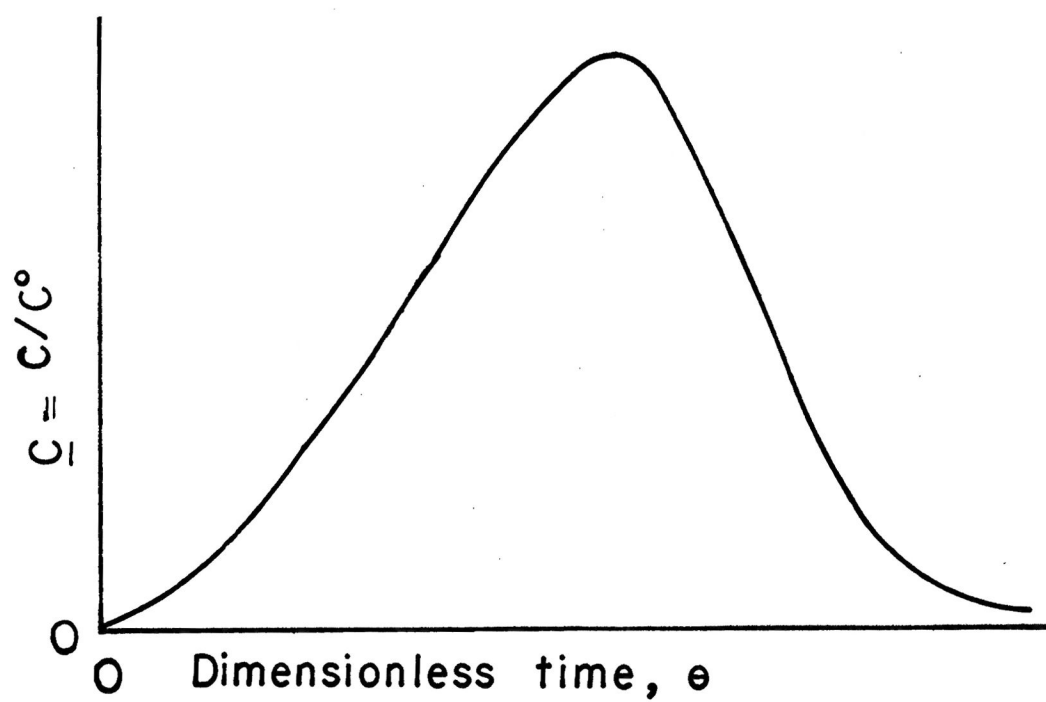


Fig.4. Typical \underline{C} -curve

response to a delta function or unit impulse input is called the C-curve. As with the F-curve, the range and domain are dimensionless. Concentrations are measured in terms of the initial concentration, C^0 , as if it were evenly distributed throughout the vessel.

$$C^0 = \int_0^{\infty} C d\theta = \frac{1}{\bar{t}} \int_0^{\infty} C dt \quad (18)$$

Time is measured in reduced units. With this choice of units

$$\int_0^{\infty} \underline{C} d\theta = 1 \quad (19)$$

Fig. 4 shows a typical C-curve. The terms F, C, I, and E were introduced by Danckwerts (29).

Correspondence between F, C, I, E. It may be shown by material balance consideration that:

$$\underline{F}(\theta) + \underline{I}(\theta) = 1 \quad (20)$$

$$\underline{C}(\theta) = \underline{E}(\theta) \quad (21)$$

$$\underline{F} = 1 - \underline{I}(\theta) = \int_0^{\theta} \underline{E}(\theta) d\theta = \int_0^{\theta} \underline{C}(\theta) d\theta \quad (22)$$

$$\underline{C}(\theta) = \underline{E}(\theta) = \frac{d\underline{F}(\theta)}{d\theta} = - \frac{d\underline{I}(\theta)}{d\theta} \quad (23)$$

The point age distribution. Let the concentration of a particular species at the entrance of a flow system be changed and

the resulting change with time at some point within the system be observed. Spalding (30) and Danckwerts (31) have shown that it is possible to deduce the age distribution of the fluid flowing through a volume element around that point from the variation of the point concentration with time.

In particular, let a step input of tracer be introduced at the inlet of the vessel, changing the concentration of tracer in the inlet stream from 0 to C_0 . At the observation point, the concentration of tracer, C_p , will increase with time from zero to a maximum and steady state value C_p^* . The plot of C_p/C_p^* versus θ can be interpreted to represent the cumulative age distribution (F-curve) at the point. If it is assumed that the fluid at the observation point is completely mixed within the sampling volume, the steady state value of the concentration C_p^* represents the total amount of fluid elements in the sampling volume and therefore the fraction $\frac{C_p}{C_p^*}$ is a true cumulative probability function such that:

$$\int_0^{\infty} \frac{d\left(\frac{C_p}{C_p^*}\right)}{d\theta} d\theta = 1 \quad (24)$$

It follows that $\frac{d\left(\frac{C_p}{C_p^*}\right)}{d\theta}$ is a density or frequency distribution function, $E_p(\theta)$, which is called the point age distribution after

Zwietering¹ (32). It should be noticed that $\underline{E}_p(\theta)$ corresponds to the r.t.d.f. for the volume element under consideration

From the definition of $\underline{E}_p(\theta)$ the point age distribution may be related to the internal age distribution of the entire vessel as

$$\underline{I}(\theta) = \frac{1}{V} \sum_{\substack{\text{all} \\ \text{points}}} \underline{E}_p(\theta) v_p \quad (25)$$

where v_p is the volume of the sampling point. As discussed by Danckwerts (31), v_p should be small compared to the system volume but large enough to contain many molecules or fluid elements.

Point age distribution has been treated by Zwietering (32) in the most general manner by requiring only that the point under consideration be defined for a particular instant of time.

However, Spalding (30) has shown that if the velocity vector at a point and the local effective diffusivity do not vary with time or concentration then

$$\int_0^{\infty} C(x,y,t) dt = \text{constant, everywhere} \quad (26)$$

This means that C_{p*} is equal to C_0 everywhere within the system for rather general and complex flow conditions within the vessel.

¹Zwietering called the point age distribution function, ϕ_p

where $\phi_p = \frac{d(\frac{C_p}{C_{p*}})}{dt}$. It has been denoted by $\underline{E}_p(\theta)$ here for the sake of consistency.

This is equivalent to saying that the structure of flow in the vessel remains constant in time. That is, at the sample point, which is fixed in both space and time, the elements of fluid being sampled are essentially all of the same nature so that sampling at a point is synonymous with sampling from a single element of fluid over a given period of time (28). Longfield and DeMaria (28) have shown that for the fluidized beds they treated, the structure of flow in the gas phase could be assumed constant in time at a fixed point in space.

Utilization of age distributions. Consider a flow vessel in which a rate process is taking place. If the rate process is linear, we can predict the performance of the vessel if we are given two pieces of information; the residence time distribution function for plain unchanging fluid passing through the vessel, and the complete description of the rate process (21).

If the rate process is non-linear, the performance of the vessel cannot be predicted from these separate pieces of information. The actual flow pattern of fluid through the vessel must be known before performance predictions can be made.

Finding the actual detailed flow pattern experimentally and then trying to interpret this information is impractical. Therefore, the approach taken when the flow pattern is needed to predict system performance is to formulate a flow model which approximates real flow and then use this model for predictive purposes. If the flow model reflects the real situation, the r.t.d.f.'s predicted by the model will closely match the r.t.d.f.'s obtained from the experimental tracer response curves

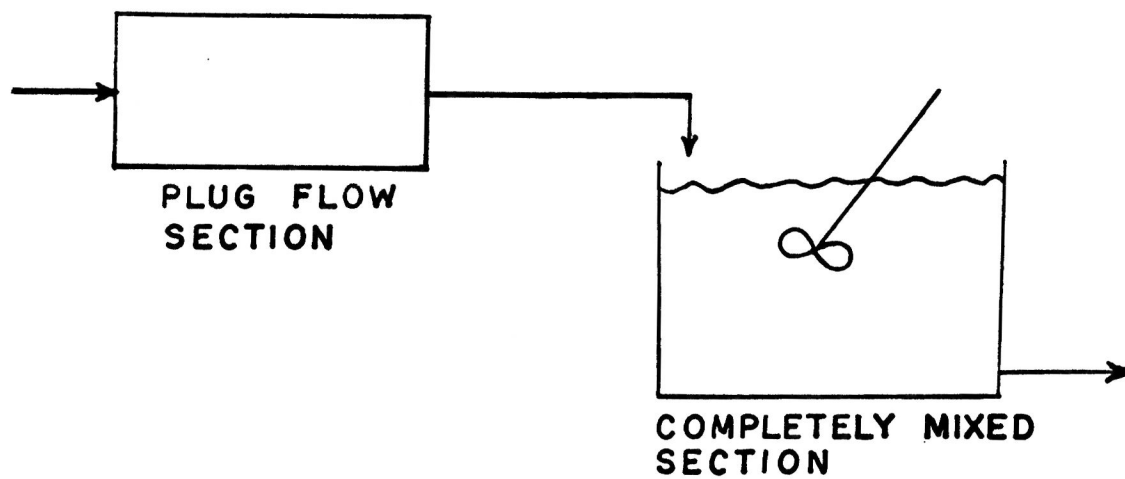
of the real vessel. This is one of the requirements in selecting a suitable model.

The requirement that r.t.d.f.'s predicted from the flow model match those obtained experimentally is not always sufficient to insure the selection of the most suitable model. For a given situation it may be possible to formulate several models for which the predicted r.t.d.f.'s match those obtained experimentally, however, none of these models may describe the actual flow behavior with sufficient accuracy. As an example, consider the two models shown in Fig. 5 which are to approximate flow conditions in a chemical reactor.

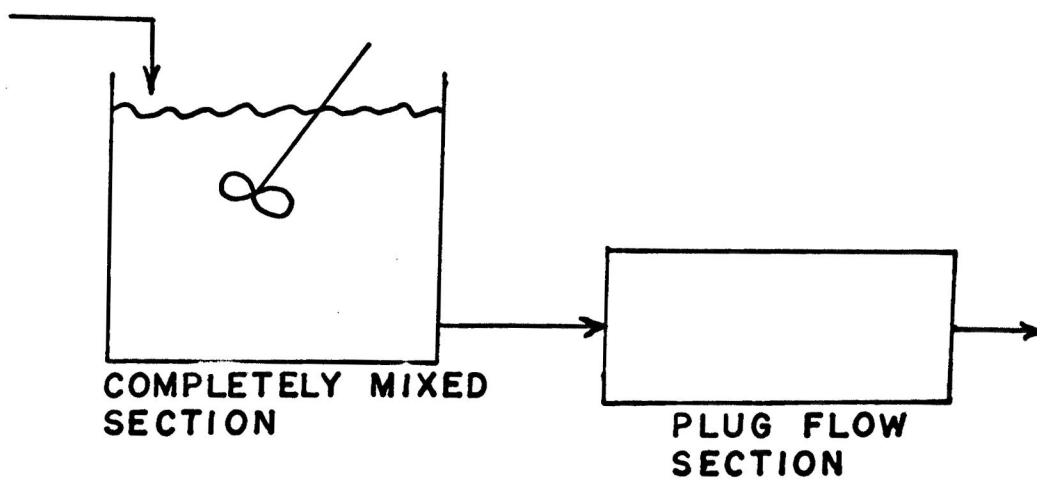
Both of these models predict the same r.t.d.f.'s. If the rate process which is to be carried out is represented by a linear rate equation, it makes no difference which model is used to predict the performance of the actual system; both will predict the same degree of completion of the process.

If, on the other hand, the rate process is represented by a non-linear rate equation, Model I predicts a different degree of completion than does Model II. In this case we must also be concerned with the degree of segregation, i.e., does micromixing, macromixing, or some combination of the two represent actual behavior.

For both models, the assumption of no micromixing or complete segregation will lead to higher predicted degree of completion than will the assumption of complete micromixing or complete lack of segregation for the higher order rate processes.



MODEL I



MODEL II

FIG. 5. MODELS OF A FLOW VESSEL

Point age distribution may be used in various ways to give a better understanding of the flow behavior and transport properties of a system. Some statistical property of the point age distribution such as the median or some moment of the distribution may be mapped as a function of space on a cross sectional drawing of the vessel (28). In the case of the example given above, such a treatment would be a valuable aid in determining which model more closely approximates the actual flow system. Treatments of this type are exemplified by Place, Ridgeway, and Danckwerts (33) in the study of air flow patterns in a spray drier and by DeMaria and Longfield (28) in the study of gas phase flow patterns in fluidized beds.

Point age distribution may also be used to calculate the degree of segregation introduced by Danckwerts (31). Zwietering (32) has given a complete discussion of the value that the concept of segregation has in the prediction of the extent of conversion in a chemical reactor.

FLOW MODELS

The use of flow models, physical or mathematical, to represent actual systems is a very fruitful approach to the prediction of system performance. The parameters of these models can be correlated with the physical properties of the fluid, vessel geometry, and flow rates. Once these correlations are found performance predictions can be made without resort to experimentation for all types of fluid processing.

Many types of models can be used to describe non-ideal flow patterns within vessels. Those which draw on the analogy between mixing in actual flow and a diffusion process are called dispersion models (21). Other models, such as the two mentioned in the preceding section visualize various flow regions connected in series or parallel. If the flow regions are not all of the same type, the model is referred to as a mixed model. Models which in no way attempt to describe the mixing mechanism other than to give the r.t.d.f. are often placed in a class called mathematical models.

Models may vary in complexity depending on the number of parameters included. As the number of parameters used increases, so does the difficulty in establishing general correlations. In general, therefore, it is best to use as few parameters as is consistent with adequate description of the system of interest.

The Dispersion Model

The assumption that concentration fluctuations are numerous and random and also small with respect to the size of the vessel leads to a diffusion type equation for representing the mixing process. Even though it is known that the concentration fluctuations are not independent for turbulent mixing, it is found that a phenomenological description gives good results for many situations (34). An eddy diffusivity or dispersion coefficient is defined so that a diffusion type equation may be used. The eddy diffusivity is then found by experiment.

One of the general forms of the dispersion model is given by

$$\frac{\partial C}{\partial t} + \text{div}(-D \text{ grad } C) + \text{div}(\bar{u}C) + \psi(C) = 0 \quad (27)$$

where D is the dispersion coefficient, \bar{u} the fluid velocity vector, C the concentration, and t is time. The first term of equation (27) represents the change in concentration with respect to time. The second term represents net outflow due to dispersion. The third term represents the velocity gradient and the fourth term represents the depletion of material caused by the progress of a rate process.

The axial dispersed plug flow model. The most often treated dispersion model is the axial dispersed plug flow model which is obtained from equation (27) by making the following assumptions:

1. Only the dispersion in the axial direction is significant.

2. The dispersion coefficient, D , is independent of position and concentration gradients.

3. The fluid flows with an average axial velocity component, \bar{U} . With these assumptions equation (27) reduces to

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2} - \bar{U} \frac{\partial C}{\partial z} - \psi(C) \quad (28)$$

Equation (28) cannot be used for design purposes unless the value of the dispersion coefficient is known. The usual way of finding the values of D is through unsteady tracer injection experiments. The tracer may be injected in the form of a unit impulse or delta function, a step function, a periodic function, or an arbitrary pulse. Whatever the form of the input, the tracer is injected uniformly over a plane normal to the direction of flow and the tracer concentration is then measured downstream from the injection point. The modification of the input signal can thus be related to the dispersion coefficient (21).

For pulse type inputs, the functional relationship between the variance of the tracer curve and the dispersion coefficient is found by solving the partial differential equation for the concentration, with D as a parameter, and then finding the variance from this theoretical expression. The dispersion coefficient for the system can then be calculated from the theoretical expression for the variance and the experimentally found variance (21).

For the experimental determination of the dispersion coefficient using a delta function input of tracer, equation (28) becomes in terms of reduced quantities and variables

$$\frac{\partial C}{\partial \theta} + \frac{\partial C}{\partial \eta} - \frac{1}{Pe} \frac{\partial^2 C}{\partial \eta^2} = \delta(\eta - \eta_0) \delta(\theta) \quad (29)$$

where

δ is the Dirac Delta function

$$\eta = \frac{z}{L}$$

L = characteristic length of the system

$$Pe = \frac{UL}{D}$$

The selection of the proper boundary conditions to be used with equation (29) has been the subject of a good deal of study. The most general set of boundary conditions is obtained by considering in the manner of Wehner and Wilhelm (35) the system to be composed of three sections in series; the mixing vessel proper of length η_1 and a for- and aft-section, a and b, both of semi-infinite extent.

This allows for a different dispersion coefficient at the inlet and outlet of the system; that is, it takes into account end effects. The system as described above is shown in Fig. 6.

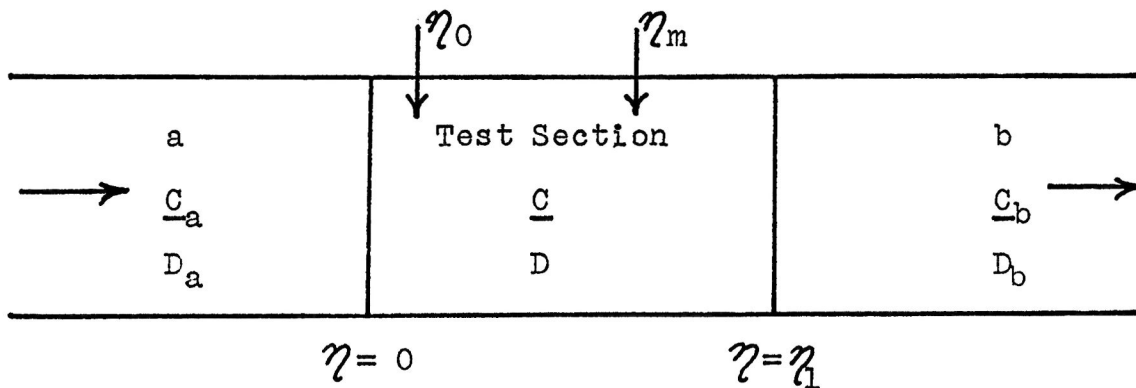


Fig. 6. General flow system of Wehner and Wilhelm (35)

The plane where the tracer is injected is denoted by η_0 and the plane of measurement is denoted by η_m . As mentioned previously, the measurement of tracer concentration at η_m in response to a unit impulse injection of tracer at η_0 yields the C-curve for the section of length $\eta_m - \eta_0$.

If the system is visualized as described above, it is necessary to solve the following set of differential equations

$$\frac{\partial \underline{C}_a}{\partial \theta} + \frac{\partial \underline{C}_a}{\partial \eta} - \frac{1}{Pe_a} \frac{\partial^2 \underline{C}_a}{\partial \eta^2} = 0 \quad \eta \leq 0 \quad (30-a)$$

$$\frac{\partial \underline{C}}{\partial \theta} + \frac{\partial \underline{C}}{\partial \eta} - \frac{1}{Pe} \frac{\partial^2 \underline{C}}{\partial \eta^2} = \delta(\eta - \eta_0) \delta(\theta) \quad 0 < \eta < 1 \quad (30-b)$$

$$\frac{\partial \underline{C}_b}{\partial \theta} + \frac{\partial \underline{C}_b}{\partial \eta} - \frac{1}{Pe_b} \frac{\partial^2 \underline{C}_b}{\partial \eta^2} = 0 \quad \eta \geq 1 \quad (30-c)$$

With initial and boundary conditions

$$\underline{C}_a(\eta, 0) = \underline{C}(\eta, 0) = \underline{C}_b(\eta, 0) = 0 \quad (31-a)$$

$$\underline{C}_a(-\infty, \theta) = \text{finite} \quad (31-b)$$

$$\underline{C}_a(0^-, \theta) = \underline{C}(0^+, \theta) \quad (31-c)$$

$$\underline{C}_a(0^-, \theta) - \frac{1}{Pe_a} \frac{\partial \underline{C}_a(0^-, \theta)}{\partial \eta} = \underline{C}(0^+, \theta) - \frac{1}{Pe} \frac{\partial \underline{C}(0^+, \theta)}{\partial \eta} \quad (31-d)$$

$$\underline{C}(\eta_1^-, \theta) - \frac{1}{Pe} \frac{\partial \underline{C}(\eta_1^-, \theta)}{\partial \eta} = \underline{C}_b(\eta_1^+, \theta) - \frac{1}{Pe_b} \frac{\partial \underline{C}_b(\eta_1^+, \theta)}{\partial \eta} \quad (31-e)$$

$$\underline{C}(\eta_1^-, \theta) = \underline{C}_b(\eta_1^+, \theta) \quad (31-f)$$

$$\underline{C}_b(+\infty, \theta) = \text{finite} \quad (31-g)$$

The boundary value problem given by equations (30) and (31) may be simplified for various specific cases. Levenspiel and Smith (36) treated the simple case of a doubly-infinite or open vessel. This is equivalent to setting $D_a = D = D_b$. In this case only equation (30-b) is needed with the initial condition $\underline{C}(\eta, 0) = 0$ and the boundary conditions given by equations (31-b) and (31-g). The characteristic length, L , is taken as the distance between the injection and measurement points.

For the case of a closed vessel, i.e., $D_a = D = D_b = 0$, only equation (30-b) is needed with the initial condition $\underline{C}(\eta, 0) = 0$ and boundary conditions

$$\underline{C}_a(0^-, \theta) = \underline{C}(0^+, \theta) - \frac{1}{Pe} \frac{\partial \underline{C}(0^+, \theta)}{\partial \eta} \quad (32-a)$$

$$\frac{\partial \underline{C}(\eta_1, \theta)}{\partial \eta} = 0 \quad (32-b)$$

This set of boundary conditions is equivalent to that introduced by Danckwerts (29) for a tubular flow reactor under steady-state conditions.

Other approximate boundary conditions have been proposed for the case of a closed vessel by Hulburt (37), Levenspiel (36), and Ahn (38).

If the unit impulse of tracer is injected upstream from section a as shown in Fig. 7, equation (30-a) is no longer needed

and only two initial conditions and four boundary conditions are required.

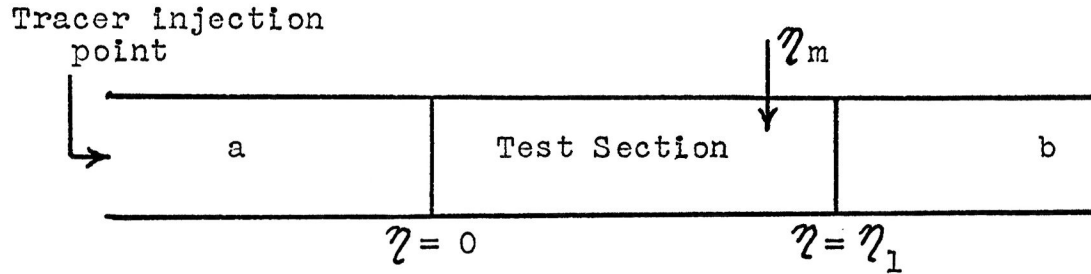


Fig. 7. General flow system with downstream tracer injection

Van der Laan (39) obtained a general solution to the boundary value problem of equations (30) and (31) in the Laplace transform plane and without inversion obtained the mean, $\bar{\theta}$, and the variance, σ^2 , of the r.t.d.f. using the relations given below.

$$\underline{C}(\eta, p) = \mathcal{L} \underline{C}(\eta, \theta) \quad (33)$$

$$\lim_{p \rightarrow 0} \left(- \frac{d \underline{C}(\eta, p)}{dp} \right) = \int_0^{\infty} \theta \underline{C}(\eta, \theta) d\theta = \bar{\theta} \quad (34)$$

$$\lim_{p \rightarrow 0} \left[\frac{d^2 \underline{C}(\eta, p)}{dp^2} - \left(\frac{d \underline{C}(\eta, p)}{dp} \right)^2 \right] = \int_0^{\infty} \theta^2 \underline{C}(\eta, \theta) d\theta - \bar{\theta}^2 = \sigma^2 \quad (35)$$

The results he obtained for the most general and various simplified cases are given in Table 1.

The methods given above for determining the dispersion coefficient depend on being able to represent the tracer injection by a delta function, a mathematical idealization which can only be approximated physically. Aris (40), Bishoff (41), and

TABLE I. EXPRESSIONS FOR THE MEAN AND THE VARIANCE OF $\zeta(\theta)$, (39)

		$\bar{\theta}$	σ^2
I		$1 + \frac{1}{Pe} [2 - (1-\alpha)e^{-Pe\eta_0} - (1-\beta)e^{-Pe(\eta_1-\eta_m)}]$	$\frac{1}{Pe^2} [2Pe + 8 + (1-\alpha)(1-\beta)e^{-Pe\eta_1} - (1-\alpha)e^{-Pe\eta_0} \{4\eta_0 Pe + 4(1+\alpha) + (1-\alpha)e^{-Pe\eta_0}\} - (1-\beta)e^{-Pe(\eta_1-\eta_m)} \{4(\eta_1-\eta_m)Pe + 4(1+\beta) + (1-\beta)e^{-Pe(\eta_1-\eta_m)}\}]$
II		$1 + \frac{1}{Pe} [2 - e^{-Pe\eta_0} - e^{-Pe(\eta_1-\eta_m)}]$	$\frac{1}{Pe^2} [2Pe + 8 + 2e^{-Pe\eta_1} - e^{-Pe\eta_0} \{4\eta_0 Pe + 4 + e^{-Pe\eta_0}\} - e^{-Pe(\eta_1-\eta_m)} \{4(\eta_1-\eta_m)Pe + 4 + e^{-Pe(\eta_1-\eta_m)}\}]$
III		$1 + \frac{1}{Pe} [\alpha + \beta]$	$\frac{1}{Pe^2} [2Pe - 2 + 2e^{-Pe} + 2(\alpha + \beta)(1 - e^{-Pe}) + 3(\alpha^2 + \beta^2) + 2\alpha\beta e^{-Pe}]$
IV		1	$\frac{2}{Pe^2} [Pe - 1 + e^{-Pe}]$
V		$1 + \frac{1}{Pe} [2 - (1-\alpha)e^{-Pe\eta_0}]$	$\frac{1}{Pe^2} [2Pe + 8 - (1-\alpha)e^{-Pe\eta_0} \{4\eta_0 Pe + 4(1+\alpha) + (1-\alpha)e^{-Pe\eta_0}\}]$
VI		$1 + \frac{1}{Pe} [2 - (1-\beta)e^{-Pe(\eta_1-\eta_m)}]$	$\frac{1}{Pe^2} [2Pe + 8 - (1-\beta)e^{-Pe(\eta_1-\eta_m)} \{4(\eta_1-\eta_m)Pe + 4(1+\beta) + (1-\alpha)e^{-Pe(\eta_1-\eta_m)}\}]$
VII		$1 + \frac{2}{Pe}$	$\frac{1}{Pe^2} [2Pe + 8]$
VIII		$1 + \frac{1}{Pe}$	$\frac{1}{Pe^2} [2Pe + 3]$

$$\alpha = \frac{Da}{D}$$

$$Pe = \frac{\bar{U}(\bar{z}_m - \bar{z}_0)}{D}$$

$$\beta = \frac{Db}{D}$$

$$\eta = \frac{\bar{z}}{\bar{z}_m - \bar{z}_0}$$

Bischoff and Levenspiel (42) have shown that this approximation need not be made if concentration measurements are taken at two points rather than one. The means and the variances of the experimental concentration curves at the two points are calculated, and the respective differences between them found. These differences can be related to the parameter Pe and thus to the dispersion coefficient. The tracer may be injected anywhere upstream from the two measurement points and may be any type of pulse input. In this case the characteristic length L is taken as the distance between the two measurement points. The first measurement point is denoted by η_0 and the second by η_m .

Aris (40) and Bischoff (41) have treated the special case shown in Fig. 8 when both measurement points are inside the test section.

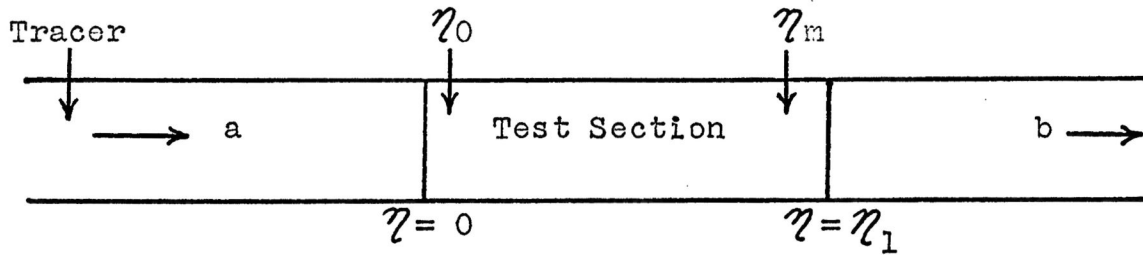


Fig. 8. Experimental setup for pulse testing

They gave the following expressions for the differences between the means and variances:

$$\Delta \bar{\theta} = \bar{\theta}_m - \bar{\theta}_0 = 1 - \frac{1-\beta}{Pe} (1 - \exp[-Pe]) \exp[Pe(\eta_m - \eta_1)] \quad (36)$$

$$\Delta \sigma^2 = \sigma_m^2 - \sigma_0^2 = \frac{2}{Pe} + \frac{1-\beta}{Pe^2} \exp[Pe(\eta_m - \eta_1)] \left\{ 4(1+\beta) \right. \\ \left. (\exp[-Pe] - 1) + 4Pe(\eta_m - \eta_1) + (1-\beta)(\exp[-2Pe] - 1) \right. \\ \left. \exp[Pe(\eta_m - \eta_1)] + 4Pe(\eta_1 - \eta_0) \exp[-Pe] \right\} \quad (37)$$

Equations (36) and (37) reduce to particularly simple forms for the case of an infinite vessel where $\beta = 1$, i.e., $D_b = D$.

$$\bar{\theta} = \bar{\theta}_m - \bar{\theta}_0 = 1 \quad (38)$$

$$\sigma^2 = \sigma_m^2 - \sigma_0^2 = \frac{2}{Pe} \quad (39)$$

Equations (38) and (39) may also be used when the second measurement point is located far enough from the end of the vessel so that end effects are negligible. Bischoff and Levenspiel (42) give design charts which permit estimation of this distance. Also, equations (36) and (37) can be used to estimate the magnitude of the end effects as represented by the second term of equation (37).

As previously mentioned, a step input or some periodic function may be used for the tracer injection. However, as pointed out by Levenspiel (21), these methods are not as convenient as pulse techniques. For a step input, there seems to be no convenient method for relating the experimental response to the dispersion coefficient as was provided by the variance with the pulse input. The use of a periodic function, such as a sine wave requires much more experimental data than does the use of a

pulse technique. Also, the working equations are usually quite complicated.

Determination of the dispersion coefficient by steady state tracer injection involves injection of an inert tracer into the fluid stream a short distance upstream from the vessel outlet and subsequent measurement of the degree of tracer diffusion upstream from the injection plane. The tracer is added continuously and the steady state values of tracer concentration as a function of position are determined.

For this situation equation (28) reduces to

$$\frac{D}{\bar{U}} \frac{d^2 C}{dz^2} - \frac{dC}{dz} = 0 \quad (40)$$

If the vessel has a sufficient length of fluid travel so that none of the tracer reaches the inlet section of the vessel, the solution of equation (40) is

$$\frac{C - C_0}{C_i - C_0} = \exp \left[- \frac{\bar{U}z}{D} \right] \quad (41)$$

where

C = tracer concentration at a distance z upstream of the injection point.

C_0 = concentration of tracer in feed stream

C_i = concentration of tracer at the injection point.

The value of \bar{U}/D can be obtained from the slope of experimental data on a plot of $\ln(C - C_0)$ vs. z .

The dispersed plug flow model. The dispersed plug flow model takes into account the dispersion in both the longitudinal and lateral directions. The parameters of this model are D_L , the axial or longitudinal dispersion coefficient and D_R , the lateral or radial dispersion coefficient. As with the axial dispersed plug flow model, the dispersion coefficients are assumed to be independent of position and concentration gradients, and the fluid is assumed to flow with an average axial velocity component, \bar{U} . With these assumptions, equation (27) becomes in rectangular coordinates

$$\frac{\partial C}{\partial t} = D_L \frac{\partial^2 C}{\partial z^2} + D_R \frac{\partial^2 C}{\partial x^2} - \bar{U} \frac{\partial C}{\partial z} - \psi(C) \quad (42)$$

where

z = longitudinal direction (direction of flow)

x = lateral direction

The method of solution is similar to that of the axial dispersed plug flow model except for modifications which keep both axial and lateral dispersion in the equations.

Additional comments on dispersion models. Only the axial dispersed plug flow model has been discussed here in detail since this is the model most frequently employed to describe mixing on distillation plates by Byfield (43), Anderson (13), and Ruckenstein (44), and was used by Gerster (15) in a general correlation for determining plate efficiency. Eddy diffusion coefficients have been measured experimentally for bubble cap and sieve plates by several investigators.

Brown (16), Stone (45), Wharton (17), Robinson (5), and Gerster (15) used steady state tracer injection techniques for bubble cap trays. Olson (46) and Barker and Self (47) used the delta function input for sieve plates and Leonard (3) and Gilbert (48) used a sinusoidal tracer input on bubble cap trays. These investigators differ in opinions as to the suitability of using a dispersion model to describe mixing on distillation plates. It is perhaps significant that no one has yet used the experimental technique which requires the fewest assumptions, i.e., the arbitrary pulse technique (or the pulse testing method) with two measurement points.

The reader is referred to Levenspiel (21) for a complete discussion of the general dispersion model and the other less general models derived from it.

It should also be noted that for the case of laminar flow, analytic solutions equivalent to the solutions obtained from the dispersion model can be obtained. Taylor (49) showed for laminar flow in round empty tubes that dispersion due to molecular diffusion and radial velocity variation may be represented by flow with a flat velocity profile equal to the actual mean velocity, U , and with an effective axial dispersion coefficient $D = R_0^2 U^2 / 48 \mathcal{D}$. (R_0 is the tube radius and \mathcal{D} the molecular diffusivity). Farrell and Leonard (50) have extended Taylor's work and presented a solution for the two dimensional convection-diffusion equation for laminar flow which makes no assumption concerning the relative importance of radial diffusion or axial convection as dispersion mechanisms.

Perfectly Mixed Tanks in Series Model

As previously mentioned, the perfectly mixed tanks in series model was first applied to distillation plates by Kirchbaum (6), who called it the pool model. This is a one parameter model, the parameter being, j , the number of tanks in series.

Only the case where all the tanks have the same volume will be considered here. A discussion of the more general case of unequal size tanks is given by Mason and Piret (51).

Determination of the parameter j . The C-curve for a single perfectly mixed tank can be found easily by a material balance;

$$C^0 V \delta(t) = vC + V \frac{dC}{dt} \quad (43)$$

In dimensionless form equation (43) becomes

$$\delta(\theta) = \underline{C} + \frac{d\underline{C}}{d\theta} \quad (44)$$

where

$$\theta = \frac{t}{\tau} = \frac{tv}{V}$$

$$\underline{C} = \frac{C}{C^0}$$

Solving equation (44) by the method of Laplace transform gives

$$\underline{C} = e^{-\theta} \quad (45)$$

The preceeding results can be generalized to the case of j perfectly mixed tanks in series as discussed by Ham and Coe (52),

MacMullin and Weber (53), Katz (54), and others (55), (56).

Consider j tanks in series, each having the same volume. A material balance around the i^{th} tank gives

$$C_{i-1} = vC_i + V_i \frac{dC_i}{dt} \quad (46)$$

The \underline{C} -curve may be found by solving the set of equations (46) for $i = 1, 2, \dots, j$ with the condition that the input to the first tank, $i = 1$, is a delta function of tracer. Solving equations (46) by the use of Laplace transforms gives (21)

$$\underline{C}_j = \frac{j^j}{(j-1)!} e^{j-1} e^{-j\theta} \quad (47)$$

where

$$\theta = \frac{tv}{V}$$

The mean and the variance are

$$\bar{\theta} = 1 \quad (48)$$

$$\sigma^2 = \frac{1}{j} \quad (49)$$

Hence, the experimental \underline{C} -curve data can be used to determine a variance σ^2 and thus j .

As with the axial dispersed plug flow model, the pulse testing technique can be used to determine j if two measurement points are used. Consider the experimental setup of Fig. 9.

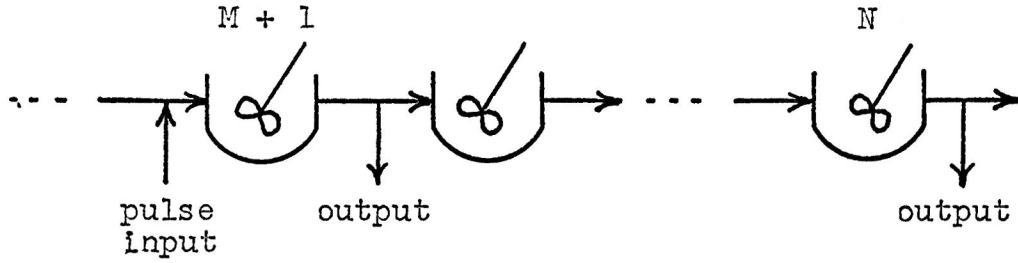


Fig. 9. Pulse testing-perfectly mixed tanks in series

Tracer concentration is measured both entering the $M + 1^{\text{th}}$ tank and leaving the N^{th} tank. Let $j = N - M$ be the number of tanks between the two measurement points. For this case (21)

$$\Delta \bar{\theta} = \bar{\theta}_N - \bar{\theta}_M = 1 \quad (50)$$

$$\Delta \sigma^2 = \sigma_N^2 - \sigma_M^2 = \frac{1}{j} \quad (51)$$

Therefore, if an arbitrary pulse of tracer is injected into a system and the mean and variance measured at two locations, the parameter, j , of the stirred tanks in series model can be determined.

Comparison with the axial dispersed plug flow model. Several authors (21), (57), (58) have discussed the similarity between the axial dispersed plug flow model and perfectly mixed tanks in series model. Various methods of comparison have been suggested. Kramers and Alberda (57) used the variance for the doubly infinite vessel which from Table 1, VII is

$$\sigma^2 \approx \frac{2}{Pe} = 2 \frac{D}{UL} \quad (52)$$

Comparing this with equation (49) gives

$$\frac{1}{j} = 2\left(\frac{D}{UL}\right) \quad (53)$$

Equation (53) extrapolates properly to $j = \infty$ as $D \rightarrow 0$, but does not extrapolate to $j = 1$ as $D \rightarrow \infty$. Levenspiel (21) has shown that the reason for the incorrect extrapolation of equation (53) as $D \rightarrow \infty$ is that the doubly infinite vessel is not the proper one to use for the comparison. The closed vessel ($D_a = D_b = 0$) must be used. Then from Table 1, IV,

$$\frac{1}{j} = 2\left(\frac{D}{UL}\right) - 2\left(\frac{D}{UL}\right) [1 - e^{-UL/D}] \quad (54)$$

This expression extrapolates properly to $j = 1$ for $D \rightarrow \infty$. For small values of $1/Pe$ it reduces to equation (53).

These comparisons are the basis for the statement that an infinite number of stirred tanks in series is equivalent to plug flow (21).

Trambouze (58) has suggested that the models be compared by matching either the C-curve maxima for the two models or by matching the C-curve at $\theta = 1$. As pointed out by Levenspiel (21), there is no unique way of matching the two models.

Mixed Models

Components of mixed models. Models which suppose the real system to consist of interconnected flow regions with various

modes of flow between and around these regions are called mixed models (21). These models may be constructed from some combination of the following regions and types of flow:

plug flow regions

perfectly mixed regions

dispersed plug flow regions

deadwater regions

bypass flow, where a fraction of the fluid bypasses the vessel or a specific flow region

recycle flow, where a fraction of the fluid leaving the vessel or specific flow region is recycled and mixed with the fluid entering the vessel or region

cross flow, where interchange, but no net flow, of fluid occurs between various flow regions.

Deadwater regions account for the fraction of fluid in the system which is relatively slow moving and for all practical purposes, stagnant (21). These regions may be defined or viewed in two ways. The first and most generally used definition is given by Levenspiel (59).

In a vessel the deadwater regions are the relatively slow moving portions of the fluid which we chose to consider to be completely stagnant. Deadwater regions contribute to the vessel volume; however we ignore these regions in determining the various age distributions.

The second definition considers that there is a slow interchange or cross flow between the deadwater regions and the active fluid passing through the vessel. Adler and Hovorka (60), Turner (61), (62), and Aris (63), (64) have illustrated this concept.

Levenspiel (21) gives the following relationship for determining the number of parameters in a mixed model

$$\begin{aligned}
 \text{Number of Parameters} = & \sum \left(\text{Flow regions in excess of one} \right) + \sum \left(\text{Flow paths in excess of one} \right) \\
 & + \sum \left(\text{zones of cross flow} \right) + \sum \left(\text{Flow regions with dispersion} \right) \\
 & - \sum \left(\text{arbitrary restrictions on flow and volume ratios} \right)
 \end{aligned} \quad (55)$$

General remarks. Many mixed models have been devised to describe various systems; for example see Oliver (9), Weber (65), Eguchi (27), Gilliland (66), Bartok, Heath, and Weiss (67), Cholette and Cloutier (68), Handlos, Kunstman, and Schissler (69), Pansing (70), and Singer, Todd, and Guinn (71). Strand (19) has proposed a model for distillation plates which considers the plate to be a single dispersed plug flow region with a fraction of the liquid by passing the plate.

A discussion of each of these proposed mixed models and the specific methods used for determining their parameters will not be given here. Let it suffice to say that the usual method of testing the aptness of the model is to compare the experimental C-curve to the C-curve predicted by the model. Shepard (72) gives a complete discussion of this subject. The predicted C-curve is usually obtained by the use of Laplace transforms. The closeness of fit of the model to the experimental data is not the only criteria which must be used in selecting the proper model. The simplest model which fits the facts and whose various regions are suggested by the real vessel should be selected. As pointed out by Levenspiel (21), ". . . an unrealistic, many-parameter model may closely fit all present data after the fact,

but may be quite unreliable for predution in a new untried situation."

Γ -Distribution Model

The models discussed in the preceeding sections are physical models in that they are based on assumptions which lead to a simplified physical picture of the actual dispersion process. In this section we consider models which may be used to predict the exit age distribution function but which in no way attempt to describe the actual mixing mechanism.

Foss (18) noted that a function of the form

$$f(\theta) = \phi \theta^{\xi} e^{-\gamma \theta} \quad (56)$$

where

ϕ , ξ , and γ are constants and

$$\theta = \frac{t}{\bar{t}}$$

appears to fit reasonably well the r.t.d.f.'s of sieve plate distillation trays. Cha (73) noted that a function which has the form of equation (56) is the density function of a Γ -distribution.

$$f(x) = \frac{1}{\gamma^p \Gamma(p)} x^{p-1} e^{-x/\gamma} \quad \begin{matrix} p > 1 \\ \gamma > 0 \\ x \geq 0 \end{matrix} \quad (57)$$

where p and γ are parameters.

Thus, the Γ -distribution model for the r.t.d.f of a system is

$$\underline{E}(\theta) = \underline{C}(\theta) = \frac{1}{\nu^p \Gamma(p)} e^{p-1} e^{-\theta/\nu} \quad (58)$$

Cha (73) has also considered a system with n mixing units. He has shown that if $\underline{E}_j(\theta)$ of the j^{th} mixing unit is given by equation (50) and if the parameters ν_j for each $\underline{E}_j(\theta)$ are equal, the exit age distribution for the system as a whole is given by

$$\underline{E}_n(\theta) = \frac{1}{\nu^{np} \Gamma(np)} e^{np-1} e^{-\theta/\nu} \quad (59)$$

Since the Γ -distribution model has not yet been compared to actual data, a simplified form of equation (58), i.e., $\nu = 1$, was compared with Olson's (46) data which were obtained by injecting a unit impulse of tracer evenly across the inlet weir of a sieve plate distillation tray and measuring the concentration immediately beyond the outlet weir. Sodium chloride was used as the tracer and concentration was measured by a conductivity cell in an unbalanced wheatstone bridge. Even though the experimental technique outlined above is perhaps not the most desirable one to use for obtaining the $\underline{C}(\theta)$ -curve, data of this nature are relatively scarce, and it was felt that the data would give some information concerning the suitability of the Γ -distribution model for predicting r.t.d.f.'s or distillation plates.

The model used is given in terms of its exit age distribution by

$$\underline{E}(\theta) = \underline{C}(\theta) = \frac{\theta^{p-1} e^{-\theta}}{\Gamma(p)} \quad (60)$$

The parameter p was determined by the method of least squares. A computer program for this purpose written for the IBM 1620 is given in the Appendix. The value of p which gave the best fit was found to be $p = 7.650$. This indicates considerable deviation from perfect mixing where p is equal to unity. A comparison of the model and that data is given in Fig. 10.

The goodness of fit indicates that it may be possible to use the one-parameter Γ -distribution model as represented by equation (60) to predict the r.t.d.f.'s for distillation plates and a reliable correlation for the parameter, p , as a function of the physical parameters of the system of the model and operating conditions may be found.

While it is true that the two-parameter model as represented by equation (50) would better fit the data, the use of such a model at this stage cannot be justified because of the limited amount of the sufficiently accurate data available.

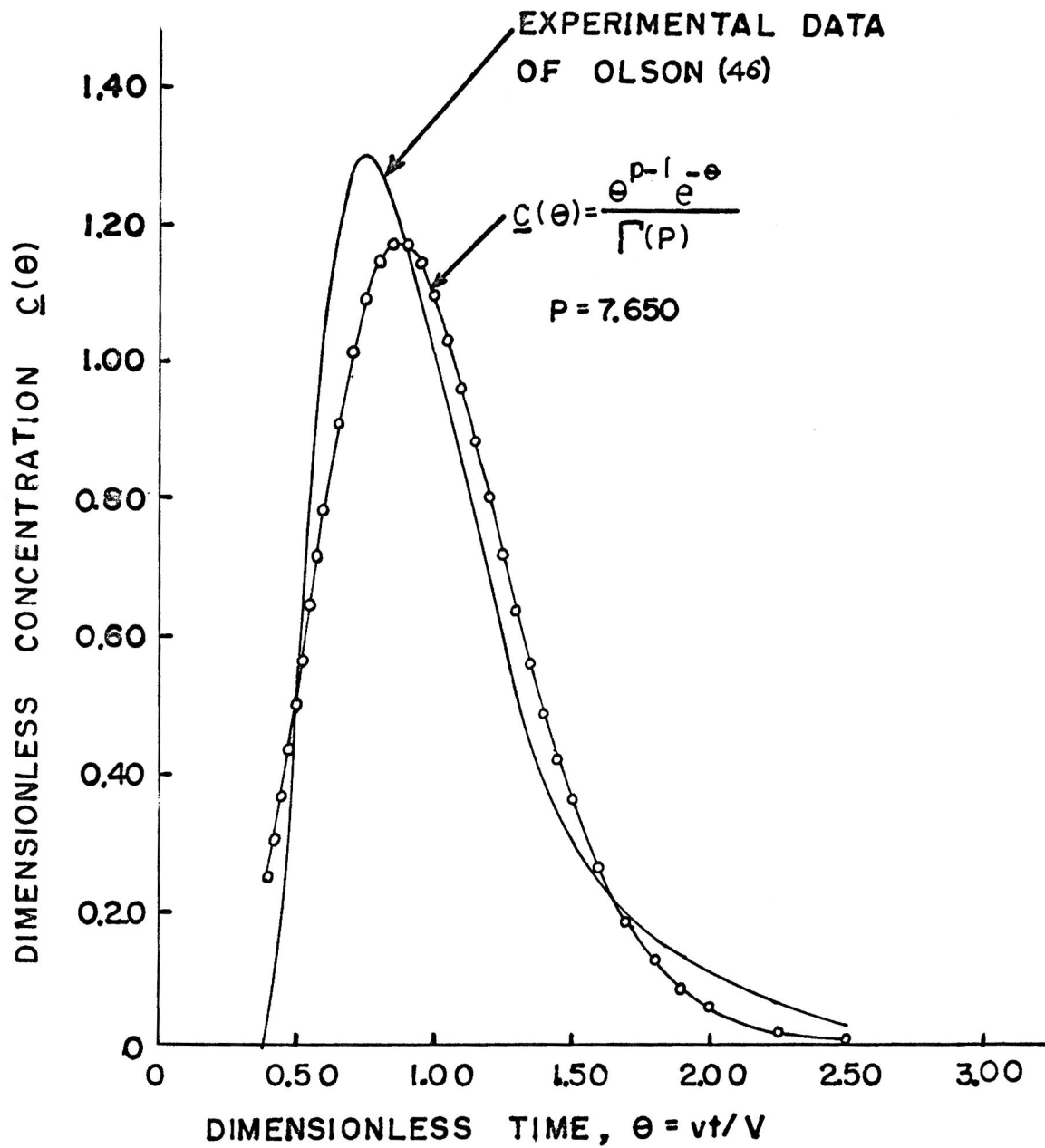


FIG.10. COMPARISON OF Γ -DISTRIBUTION MODEL
WITH EXPERIMENTAL DATA

PROPOSED INVESTIGATION OF LIQUID MIXING AND DISPERSION ON DISTILLATION PLATES

Introduction

In order to point out the need for further investigation of liquid mixing on distillation plates, we need only review the conclusions and recommendations of several investigators of this phenomenon. Leonard (3) concludes that the r.t.d.f.'s of bubble cap trays are roughly those which would be observed for a series of n perfect mixing tanks. However, he points out that the frequency response data that he obtained should be reworked to yield more accurate r.t.d.f.'s. He also points out the need for direct observation of the degree of vertical mixing, examination of the froth for stagnant pockets, and a general picture of a multi-dimensional froth movement on active trays.

Gerster (15) and Robinson (5), from their studies of bubble cap trays, conclude that the axial dispersed plug flow model gives an adequate description of the liquid mixing. Olson (46) however, from his study of sieve plates, concludes that this model is not adequate.

Strand (19) has shown that the bubble tray design method proposed by the American Institute of Chemical Engineers (20), which uses the axial dispersed plug flow model to describe fluid mixing, is unsatisfactory for unusual combinations of system properties and vapor and liquid flows. One suggested reason for deficiencies of the A.I.Ch.E method and of other correlations in

the literature is that they do not provide a means for assessing the effects of liquid and/or vapor bypassing. It is shown that the A.I.Ch.E. method could be substantially improved by including a liquid bypassing effect. Strand also suggests that it may be fruitful to consider the tray to be non-uniform, both in the contacting achieved on various parts of the tray and in the degree of liquid mixing. This supports Leonard's (3) observation that more information is needed concerning the nature of flow on distillation plates.

The pulse testing technique has not been used for determining the r.t.d.f.'s of distillation plates or for testing the suitability of proposed models. Either the injection of a perfect unit impulse or a perfect step input of tracer have been assumed in most previous investigations. This presents some question as to the reliability of existing data.

Areas of Investigation and Experimental Techniques

Any future investigation of liquid mixing on distillation plates should have in part the following objectives:

1. To obtain detailed information concerning the local froth and liquid flow behavior.
2. To assess the suitability of various models for predicting the residence time distribution functions.

The achievement of the first objective would give a better understanding of the actual mixing mechanism and thereby serve as the basis for the selection of realistic physical models.

This objective can be attained by experimentally measuring point age distributions and then mapping some property of these distributions such as the mean as a function of position. A mapping of this nature would detect the existence of various flow regions such as deadwater, plug flow, or perfectly mixed regions, and thus provide the basis for model selection.

It is suggested that a radioactive material be used as the tracer for both point age distribution studies and residence time distribution measurements. Small amounts of radioactive material are easily detected. This means that a very small amount of radioactive tracer could be added to the liquid stream causing little fluctuation in flow rate. An additional advantage of using a radioactive tracer is that samples need not be withdrawn from the liquid stream.

For point age studies, tracer concentration could be monitored at several points on the tray simultaneously by using several scintillation probes and a multichannel recorder. Part of the instrumentation used by King (74) in the study of a particale mixing in a liquid-solid fluidized system would be ideally suited for this purpose. The scintillation probe he used was a Nuclear-Chicago Model DS8 scintillation detector. This unit consists of a transistorized preamplifier and a 10-stage photomultiplier with cesium dynodes and is housed in an aluminum container 9.25 inches long and 1 inch in diameter. A 3 mm. diameter by a 10 cm. long needle probe is used with the basic detector. This beta-gamma sensitive needle probe is sensitive only at the tip.

The second objective can be attained by experimentally measuring the r.t.d.f.'s and comparing this data with the r.t.d.f.'s predicted by the various models. It is suggested that the pulse testing technique with two measurement points be mainly used for experimentally determining the r.t.d.f.'s. Methods for converting pulse data to unit impulse data are outlined in the following section.

Initial studies should be made using the air water system. These materials are relatively cheap and need not be recovered. Other systems should be used as experimental techniques are perfected.

The investigation outlined above should be carried out using trays of various designs and covering a wide range of operating conditions. The majority of the work should be done with sieve and valve trays. In recent years these types of trays have been superseding conventional bubble cap trays as the preferred contacting device for many separation processes (75), however, most of the data in the published literature are for bubble cap trays.

One approach to the study of the effect of tray design on liquid mixing and dispersion would be to study first a tray with only one perforation, valve, or bubble cap and then observe the effect of adding additional perforations, valves, or bubble caps.

Methods of Data Analysis and Presentation

For many models which might be used to describe a flow system, it is most convenient to relate the parameters of the

models to the C-curve or to some property of the C-curve such as the variance. For example, the parameter, p , of the Γ -distribution model was found by fitting the experimental C-curve to the predicted C-curve.

It may also be desirable to compute the transfer function of the system; the transfer function being defined as the ratio of the output signal to the input signal in the Laplace transform plane. The transfer function may be obtained from the flow model of the system. As discussed by Law and Bailey (76) and Hougén and Walsh (77), it may also be found from frequency response and pulse response data approximately and empirically.

Since it has been suggested in the previous section that the pulse testing technique be used to obtain experimental data, it is appropriate that methods of obtaining the unit impulse response and the frequency response of a system from pulse testing data be discussed here.

The conversion of pulse testing data to frequency response is based on the fact that the transfer function can be expressed as

$$G(j\omega) = \frac{\bar{F}[C_o(t)]}{\bar{F}[C_i(t)]} \quad (61)$$

where

\bar{F} is the Fourier transform operator

$$j = \sqrt{-1}$$

ω = frequency, rad./unit time

$C_i(t)$ = input signal; in this case, concentration at first measurement point

$C_o(t)$ = output signal; in this case, concentration at second measurement point

t = time.

The Fourier transform of a function of time is defined as

$$F(j\omega) = \mathcal{F}[f(t)] = \int_0^{\infty} f(t) e^{-j\omega t} dt \quad f(t) = 0, t < 0 \quad (62)$$

The integral of equation (62) can be divided into a real and an imaginary part, where the real part is

$$R[F(j\omega)] = \int_0^{\infty} f(t) \cos \omega t dt \quad (63)$$

and the imaginary part is

$$I[F(j\omega)] = - \int_0^{\infty} f(t) \sin \omega t dt \quad (64)$$

Therefore the transfer function can be expressed as

$$G(j\omega) = \frac{\int_0^{\infty} C_o(t) \cos \omega t dt - j \int_0^{\infty} C_o(t) \sin \omega t dt}{\int_0^{\infty} C_i(t) \cos \omega t dt - j \int_0^{\infty} C_i(t) \sin \omega t dt} \quad (65)$$

Huss and Donegan (78) have presented tables for the evaluation of the integrals appearing in equation (65). The desired

information is then given by

$$\text{amplitude ratio} = |G(j\omega)| \quad (66)$$

$$\text{phase lag} = \text{argument of } G(j\omega) \quad (67)$$

As indicated by Huss and Donegan (79), the time response to a unit impulse, $\underline{C}(t)$, can be related to the frequency response by

$$\underline{C}(t) = \frac{2}{\pi} \int_0^{\infty} R[G(j\omega)] \cos \omega t \, d\omega \quad (68)$$

Huss and Donegan (79) have given a complete discussion of the evaluation of the integral of equation (68). The method they suggest involves the approximation of $R[G(j\omega)]$ by a staircase type of function having equal frequency intervals and of such height that the area under each step of the staircase function equals the area under that portion of the $R[G(j\omega)]$ curve within the interval.

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NOTATION

C	= concentration, m/l^3
C_i	= molar concentration of component i , $\frac{\text{mols.}}{l^3}$
\underline{C}	= dimensionless concentration
$\underline{C}(\theta)$	= dimensionless response curve to a unit impulse input, a function of θ
C^0	= mean concentration of pulse of tracer if uniformly distributed in experimental section of vessel, m/l^3
C_0	= concentration of tracer in inlet stream, m/l^3
C_p	= concentration of tracer at a point, m/l^3
C_p^*	= steady state concentration at a point, m/l^3
D	= dispersion coefficient, l^2/t
D_L	= axial dispersion coefficient, dispersed plug flow model, l^2/t
D_R	= radial dispersion coefficient, dispersed plug flow model, l^2/t
$\underline{E}(\theta)$	= exit age distribution function, a function of θ
$\underline{E}(t)$	= exit age distribution function, a function of t
F	= Fourier transform
\overline{F}	= Fourier transform operator
$\underline{F}(\theta)$	= dimensionless response curve to a step input of tracer, a function of θ
$\underline{F}(t)$	= dimensionless response curve to a step input of tracer, a function of t
\overline{g}	= gravitational acceleration, l/t^2

\bar{g}_i	= total body force per unit mass of component i, $1/t^2$
$G(j\omega)$	= transfer function, a function of $j\omega$
$\underline{I}(\theta)$	= internal age distribution function, a function of θ
$\underline{I}(t)$	= internal age distribution function, a function of t
j	= parameter in the perfectly mixed tanks in series model, or the number defined by $j^2 = -1$
\bar{J}_i	= molar flux of component i, $\frac{\text{mols.}}{t l^2}$
$\bar{J}_i^{(1)}$	= laminar contribution to molar flux of component i, $\frac{\text{mols.}}{t l^3}$
$\bar{J}_i^{(t)}$	= turbulent contribution to molar flux of component i, $\frac{\text{mols.}}{t l^3}$
L	= characteristic length of system, l
\mathcal{L}	= Laplace transform operator
n	= parameter of the Γ -distribution model of equation (59)
p	= a complex variable
p	= parameter of the Γ -distribution model
p	= pressure in equation (5), m/l^2
Pe	= Peclet number, $(\frac{\bar{U}L}{D})$
\bar{q}	= energy flux, m/t^3
R_i	= molar rate of production of component i, $\frac{\text{mols.}}{t l^3}$
t	= time, t
\bar{t}	= mean residence time, t
\bar{u}	= velocity, l/t
\bar{U}	= mean velocity in axial direction, l/t

\hat{U}	= internal energy per unit mass, l^2/t^2
v	= volumetric flow rate, l^3/t
v_p	= volume of point, l^3
V	= vessel volume, l^3
x	= lateral distance, l
z	= axial distance, l

Greek letters

α	= dimensionless constant
β	= dimensionless constant
γ	= dimensionless constant
$\delta(\theta)$	= Dirac delta function, a function of θ
$\delta(\eta-\eta_0)$	= Dirac delta function, a function of
η	= dimensionless distance
η_0	= tracer injection point or first measurement point
η_m	= measurement point or second measurement point
η_1	= outlet of test section
θ	= dimensionless time
$\bar{\theta}$	= mean of the residence time distribution function
μ	= viscosity, m/lt
ν	= parameter of the Γ -distribution model
ξ	= dimensionless constant
π	= 3.1416
$\overline{\pi}$	= pressure tensor, m/t^2l
ρ	= fluid density, m/l^3
ρ_i	= mass concentration of component i , m/l^3

σ^2	= variance of residence time distribution function
$\bar{\tau}$	= shear stress tensor, m/t^2l
$\bar{\tau}(l)$	= laminar contribution to shear stress tensor, m/t^2l
$\bar{\tau}(t)$	= turbulent contribution to shear stress tensor, m/t^2l
β	= dimensionless constant
$\psi(C)$	= rate of production, a function of concentration
ω	= frequency, $rad./t$

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APPENDIX

The FORTRAN program given below can be used to determine the parameter, p , of the Γ -distribution model as represented by equation (60) from experimental values of $\underline{Q}(\theta)$.

```

E CURVE FITTING BY LEAST SQUARE
  DIMENSION X(40),E(40),DES(4),P(4),EX(4),GE(1),GMP(4)
  1 FORMAT (E10.4,E10.4,E10.4,E10.4,E10.4,E10.4)
  2 FORMAT (I3)
  READ 2, NC
  DO 20 K=1,NC
20 READ 1, X(K),E(K)
  B1=-.57669867
  B2=-1.6955437
  B3=-.84224555
  B4=-.81838432
  B5=-.48706792
  B6=-.23374017
  8 READ 1, P(1),P(2),P(3),P(4)
  DO 100 I=1,4
  M=P(I)
  U=M
  Y=P(I)-U
  GME=1.+B1*(1.+B2*(1.+B3*(1.+B4*(1.+B5*(1.+B6*Y)*Y)*Y)*Y)*Y
  PM=1.
  PU=P(I)
  L=M-1
  DO 10 J=1,L
  PU=PU-1.
  10 PM=PM*PU
100 GMP(I)=PM*GME
  DES(1)=0.
  DES(2)=0.
  DES(3)=0.
  DES(4)=0.
  DO 30 K=1,NC
  DO 40 I=1,4
  EX(I)=(P(I)/GMP(I))*((P(I)*X(K))**(P(I)-1.))*EXP(-P(I)*X(K))
  PUNCH 1, X(K),EX(I),E(K)
  40 DES(I)=DES(I)+(E(K)-EX(I))*(E(K)-EX(I))
  30 CONTINUE
  PRINT 1, DES(1),DES(2),DES(3),DES(4)
150 PAUSE
  GO TO 8
  END

```


LIQUID MIXING ON DISTILLATION PLATES

by

JOHN LEROY JOHNSON

Ch.E., The University of Cincinnati, 1961

AN ABSTRACT OF A MASTER'S REPORT

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Approved by:

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Physical and mathematical models which have been or might be used to describe liquid mixing and dispersion in flow systems in general, and on distillation plates in particular, are presented. Experimental methods for determining the parameters of these flow models are also discussed.

A mathematical model which has not previously appeared in the literature, the Γ -distribution model, is presented. The residence time distribution predicted by the Γ -distribution model is given as

$$\underline{E}(\theta) = \frac{\theta^{p-1} e^{-\theta/\nu}}{\nu^p \Gamma(p)}$$

After setting the parameter, ν , equal to unity, the residence time distribution of the Γ -distribution model is compared to an experimentally determined residence time distribution of a sieve plate distillation tray. The predicted distribution is found to compare reasonably well with the experimentally determined distribution for the parameter, p , equal to 7.65.

A proposed investigation of liquid mixing and dispersion on distillation plates is given. It is suggested that point age distributions and residence time distributions be experimentally determined through the use of radioactive tracer techniques.